

PROCESS FOR DECREASING CONTENT OF PARTICULATE MATERIAL  
CONTAINED IN EXHAUST GAS FROM LEAN BURN ENGINE OR THE LIKE

BACKGROUND OF THE INVENTION

FIELD OF THE INVENTION

The present invention relates to a process for decreasing the content of a particulate material contained in an exhaust gas from a lean burn engine or the like.

It is known that this type of the particulate material has a double-layer structure comprising a carbon solid fraction (soot) and a soluble organic fraction (SOF) covering the carbon solid fraction.

DESCRIPTION OF THE RELATED ART

There is a conventionally known process for decreasing the content of a particulate material contained in an exhaust gas, for example, from a diesel engine or the like, in which a ceramic filter (diesel particulate filter) is placed into an exhaust pipe to collect the particulate material in the filter, and when an amount of the particulate material collected reaches a predetermined value, the temperature of the exhaust gas is raised by an engine control, thereby burning the particulate material.

However, the conventionally known process has the following problems: an exhausting resistance is increased by the ceramic filter, and hence an engine output cannot be sufficiently utilized; the engine control for raising the temperature of the exhaust gas is complicated; and the filter temperature may exceed 1,000°C in some cases during burning of the particulate material, resulting

in damage to the ceramic filter.

#### SUMMARY OF THE INVENTION

Accordingly, it is an object of the present invention to provide a process for decreasing the content of a particulate material, which is capable of continuously converting the particulate material by the oxidization thereof to remarkably decrease the content thereof, and of suppressing an increase in exhausting resistance to a low level.

To achieve the above object, according to the present invention, there is provided a process for decreasing the content of a particulate material contained in an exhaust gas from a lean burn engine, comprising the steps of: generating plasma in the exhaust gas discharged from the lean burn engine and containing the particulate material, to thereby produce a plurality of  $O(^1D)$  radicals and subsequently produce a plurality of per-hydroxide excited species; and oxidizing the particulate material by the per-hydroxide excited species.

When the plasma is generated in the exhaust gas, the following process is provided:  $O(^1D)$  radicals and ozone excited species are produced by the collision between electrons released from electrodes and oxygen molecules  $\rightarrow$  hydroxide excited species are produced by the reaction of  $O(^1D)$  radicals and ozone excited species with water  $\rightarrow$  per-hydroxide excited species  $HOO^*$  having a strong oxidizing force are produced by the reaction of the hydroxide excited species with oxygen. The production of the

per-hydroxide excited species is carried out independent of the temperature of the exhaust gas. The particulate material is oxidized and converted by the per-hydroxide excited species. The conversion of the particulate material by the oxidization is conducted continuously and at a relatively low temperature.

A plasma generating device has a good gas permeability, as compared with a filter, so that even if the plasma generating device is incorporated in an exhaust pipe, an increase in exhausting resistance is suppressed to a low level. Therefore, an influence cannot be exerted to an engine output.

If an intensity  $E$  of electric field is set at a value equal to or larger than  $3.0 \text{ kV/mm}$  and a power density  $D_w$  is set at a value equal to or larger than  $1 \text{ W/cm}^3$  in the plasma generating conditions, the amount of released high-energy electrons existing in the plasma is increased, and hence the production of the per-hydroxide excited species is efficiently conducted, whereby the conversion of the particulate material by the oxidization can be enhanced. However, if at least one of requirements of  $E \geq 3.0 \text{ kV/mm}$  and  $D_w \geq 1 \text{ W/cm}^3$  is lacked, the above-described effect cannot be obtained.

Further, if the surface of at least one of opposed electrodes which is opposed to the other electrodes is covered with a dielectric, the entire surface of the dielectric is uniformly charged upon application of a voltage. Therefore, the entire space between the dielectric and the other electrode opposed to the dielectric constitutes a plasma space, so that the frequency

of random contact between the particulate material and the per-hydroxide excited species can be increased to enhance the conversion of the particulate material by the oxidization. If opposed surfaces of the opposed electrodes are covered with dielectrics, respectively, an increase in the applied voltage is provided by an increase in impedance, but it is possible to provide a converting performance equivalent to that in the case where the one surface is covered, by setting the plasma generating conditions as described above.

According to the present invention, there is also provided a process for decreasing the content of a particulate material contained in an exhaust gas from a lean burn engine or the like and comprising a carbon solid fraction and a soluble organic fraction covering the carbon solid fraction, the process comprising the steps of: subjecting the soluble organic fraction to the reaction with oxygen under the presence of a catalyst to oxidize the soluble organic fraction; generating plasma in the exhaust gas to produce plasma excited species; and oxidizing the carbon solid fraction by the plasma excited species.

The soluble organic fraction usually comprises a polycyclic aromatic hydrocarbon and the like. A large amount of oxygen is contained in the exhaust gas from the lean burn engine. Therefore, if the catalyst is present, oxygen and the soluble organic fraction are brought into contact with each other on the catalyst, whereby the soluble organic fraction is oxidized and removed. This reaction occurs even at a low temperature.

When the plasma is generated in the exhaust gas, the plasma

excited species are produced by the collision between electrons released from electrodes and gas molecules. The plasma excited species include large numbers of O(<sup>1</sup>D) radicals, hydroxide excited species OH\*, per-hydroxide excited species HOO\* and the like each having a strong oxidizing force, and hence the carbon solid fraction is oxidized by the plasma excited species. The carbon solid fraction is liable to be electrically charged in the plasma, so that there is generated a phenomenon that the carbon solid fraction is oxidized efficiently in a state in which they have been collected electrostatically on the electrodes. The significance of the preliminary removal of the soluble organic fraction from the surface of the particulate material resides in this point. The conversion of the carbon solid fraction by the oxidization as described above is conducted continuously and moreover at a relatively low temperature.

A plasma generating device has a good gas permeability, as compared with a filter, so that even if the plasma generating device is incorporated in an exhaust pipe, an increase in exhausting resistance is suppressed to a low level. Therefore, an influence cannot be exerted to an engine output.

At least one selected from the group consisting of Pt, Pd, Rh, Cu, Ag and Au is used as the catalyst.

If an intensity E of electric field is set at a value equal to or larger than 3.0 kV/mm and a power density Dw is set at a value equal to or larger than 1 W/cm<sup>3</sup> in the plasma generating conditions, the amount of released high-energy electrons existing in the plasma is increased, and hence the production of the plasma

excited species is efficiently conducted, whereby the conversion of the carbon solid fraction by the oxidization can be enhanced. However, if at least one of requirements of  $E \geq 3.0$  kV/mm and  $D_w \geq 1$  W/cm<sup>3</sup> is lacked, the above-described effect cannot be obtained.

Further, if the surface of at least one of opposed electrodes which is opposed to the other electrodes is covered with a dielectric, the entire surface of the dielectric is uniformly charged upon application of a voltage. Therefore, the entire space between the dielectric and the other electrode opposed to the dielectric constitutes a plasma space, so that the frequency of random contact between the carbon solid fraction and the plasma excited species can be increased to enhance the conversion of the carbon solid fraction by the oxidization. If opposed surfaces of the opposed electrodes are covered with dielectrics, respectively, an increase in the applied voltage is brought about with an increase in impedance, but it is possible to provide a converting performance equivalent to that in the case where the one surface is covered, by setting the plasma generating conditions as described above.

Further, according to the present invention, there is provided a process for decreasing the content of a particulate material contained in an exhaust gas from a lean burn engine or the like, comprising the steps of: generating plasma in the exhaust gas discharged from the lean burn engine and containing the particulate material to produce a plurality of nitrogen dioxide molecules and a plurality of ozone molecules; and oxidizing the

particulate material by the nitrogen dioxide molecules and the ozone molecules.

When the plasma is generated in the exhaust gas, the following process is provided: oxygen atom excited species are produced by the collision between released electrons and oxygen molecules → ozone is produced by the reaction of the oxygen atom excited species with oxygen molecules → nitrogen dioxide (NO<sub>2</sub>) is produced by the reaction of ozone with nitrogen monoxide (NO). The productions of the nitrogen dioxide and ozone are carried out independent of the temperature of the exhaust gas. The particulate material PM is oxidized and converted based on the reaction represented by PM + NO<sub>2</sub> + O<sub>3</sub> → CO<sub>2</sub> + H<sub>2</sub>O + N<sub>2</sub>. The conversion of the particulate material by the oxidization is conducted continuously and moreover at a relatively low temperature.

A plasma generating device has a good gas permeability, as compared with a filter, so that even if the plasma generating device is incorporated in an exhaust pipe, an increase in exhausting resistance is suppressed to a low level. Therefore, an influence cannot be exerted to an engine output.

Yet further, according to the present invention, there is provided a process for decreasing the content of a particulate material contained in an exhaust gas from a lean burn engine, comprising the steps of: generating plasma in the exhaust gas discharged from the lean burn engine and containing the particulate material to produce a plurality of nitrogen dioxide molecules and a plurality of ozone molecules; oxidizing the particulate material

by the nitrogen dioxide molecules and the ozone molecules; and subjecting the particulate material to the reaction with the nitrogen dioxide molecules and the ozone molecules in the presence of a catalyst to oxidize the particulate material.

If the catalyst is used as described above, the above-described reaction, namely, the reaction represented by  $PM + NO_2 + O_3 \rightarrow CO_2 + H_2O + N_2$  can be promoted to greatly increase the decrement rate of the particulate material.

At least one selected from the group consisting of Pt, Pd, Rh, Cu, Ag and Au is used as the catalyst.

If an intensity  $E$  of electric field is set at a value equal to or larger than 3.0 kV/mm and a power density  $Dw$  is set at a value equal to or larger than 1 W/cm<sup>3</sup> in the plasma generating conditions, the amount of released high-energy electrons existing in the plasma is increased, and hence the productions of the nitrogen dioxide and ozone are efficiently conducted, whereby the conversion of the carbon solid fraction by the oxidization can be enhanced. However, if at least one of requirements of  $E \geq 3.0$  kV/mm and  $Dw \geq 1$  W/cm<sup>3</sup> is lacked, the above-described effect cannot be obtained.

Further, if the surface of at least one of opposed electrodes which is opposed to the other electrodes is covered with a dielectric, the entire surface of the dielectric is uniformly charged upon application of a voltage. Therefore, the entire space between the dielectric and the other electrode opposed to the dielectric constitutes a plasma space, so that the frequency

of random contact of the particulate material with the nitrogen dioxide and ozone can be increased to enhance the conversion of the carbon solid fraction by the oxidization. If opposed surfaces of the opposed electrodes are covered with dielectrics, respectively, an increase in the applied voltage is provided by an increase in impedance, but it is possible to provide a converting performance equivalent to that in the case where the one surface is covered, by setting the plasma generating conditions as described above.

#### BRIEF DESCRIPTION OF THE DRAWINGS

Fig.1 is an illustration of a first embodiment of an equipment for a particulate material content decreasing test.

Fig.2 is a diagram of a plasma generating device.

Fig.3 is a graph showing amounts of a particulate material before and after a plasma treatment.

Fig.4 is an illustration of a second embodiment of an equipment for a particulate material content decreasing test.

Fig.5 is a graph showing amounts of a particulate material before and after a catalytic reaction and after a plasma treatment for Example 4.

Fig.6 is a graph showing amounts of a particulate material before and after a catalytic reaction and after a plasma treatment for Example 5.

Fig.7 is an illustration of a third embodiment of an equipment for a particulate material content decreasing test.

Fig.8 is a graph showing amounts of a particulate material

before and after a plasma treatment and after a catalytic reaction for Example 6.

Fig.9 is a graph showing amounts of a particulate material before and after a plasma treatment and after a catalytic reaction for Example 7.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

##### (Embodiment I)

In an equipment 1 for a particulate material content-decreasing test shown in Fig.1, a commercially available diesel generator 2 is selected as a source of generating an exhaust gas containing a particulate material. A first flow rate regulating valve 4<sub>1</sub> is mounted in an exhaust pipe 3 of the diesel generator 2. In the exhaust pipe 3, a conduit 5 is connected at one end between the diesel generator 2 and the first flow rate regulating valve 4<sub>1</sub>. A second flow rate regulating valve 4<sub>2</sub>, a heater 6, a plasma generating device PG and a flow-meter 8 are mounted in the exhaust pipe 3 sequentially in the named order from the side of the exhaust pipe 3. The other end of the conduit 5 opens to the atmosphere.

In the conduit 5, a first branch pipe 9<sub>1</sub> is connected at one end between the heater 6 and the plasma generating device PG, and at the other end to a first port p<sub>1</sub> of a three-way valve 10. In the conduit 5, a second branch pipe 9<sub>2</sub> is also connected at one end between the plasma generating device PG and the flow-meter 8, and at the other end to a second port p<sub>2</sub> of the three-way valve 10. Another conduit 11 is connected at one end to a third port

$p_3$  of the three-way valve 10, and at the other end to a suction port of a suction pump 12. A filter 13 is mounted in the conduit 11.

The diesel generator 2 is EXT12D made by Honda Motor Co., Ltd., and the specification thereof is as follows: the type of an engine: a water-cooled 3-cylinder and 4-cycle diesel engine; a total displacement: 1061 cc; a fuel used: diesel light oil; a rated output: 12 kVA. The filter 13 is a Teflon-coated filter made by German Laboratory, and has a mesh size of 0.3  $\mu\text{m}$  mesh.

Referring to Fig.2, the plasma generating device PG includes a plurality of first to sixth electrodes 14<sub>1</sub> to 14<sub>6</sub> made of a metal, each of which is formed into a plate shape in the illustrated embodiment. The first to sixth electrodes 14<sub>1</sub> to 14<sub>6</sub> are mounted in a housing 15 (see Fig.1) so that they are parallel to a direction A of flowing of an exhaust gas, and the adjacent electrodes 14<sub>1</sub> and 14<sub>2</sub>; 14<sub>2</sub> and 14<sub>3</sub>; 14<sub>3</sub> and 14<sub>4</sub>; 14<sub>4</sub> and 14<sub>5</sub>; 14<sub>5</sub> and 14<sub>6</sub> are opposed to each other. The surface of the first electrode 14<sub>1</sub> located at one end, which is opposed to the second electrode 14<sub>2</sub>, is a non-coated metal surface, but the surface of the second electrode 14<sub>2</sub>, which is opposed to the first electrode 14<sub>1</sub>, is coated entirely with a dielectric 16. The arrangement relationship between the opposed surfaces of the first and second electrodes 14<sub>1</sub> and 14<sub>2</sub> applies to those between the second and third electrodes 14<sub>2</sub> and 14<sub>3</sub>; between the third and fourth electrodes 14<sub>3</sub> and 14<sub>4</sub>; between the fourth and fifth electrodes 14<sub>4</sub> and 14<sub>5</sub>; and between the fifth and sixth electrodes 14<sub>5</sub> and 14<sub>6</sub>. The first, third and fifth

electrodes 14<sub>1</sub>, 14<sub>3</sub> and 14<sub>5</sub> are connected to a power source 18 through lead wires 17, while the second, fourth and sixth electrodes 14<sub>2</sub>, 14<sub>4</sub> and 14<sub>6</sub> are earthed through lead wires 19.

If the plasma generating device PG is constructed as described above, the entire surface of each of the dielectrics 16 is uniformly charged upon application of a voltage, and hence the entire space between each of the dielectrics 16 and each of the electrodes 14<sub>1</sub> to 14<sub>6</sub> which is opposed thereto constitutes a plasma space Pp. Namely, the plasma spaces Pp are formed between the first and second electrodes 14<sub>1</sub> and 14<sub>2</sub>; between the second and third electrodes 14<sub>2</sub> and 14<sub>3</sub>; between the third and fourth electrodes 14<sub>3</sub> and 14<sub>4</sub>; between the fourth and fifth electrodes 14<sub>4</sub> and 14<sub>5</sub>; and between the fifth and sixth electrodes 14<sub>5</sub> and 14<sub>6</sub>. That is, each of the plasma spaces Pp is defined by the opposed electrodes.

Each of the first to sixth electrodes 14<sub>1</sub> to 14<sub>6</sub> is formed of a stainless steel (e.g., JIS SUS316), and has a size of 20 mm long, 50 mm wide and 1.0 mm thick, with a transverse side extending along the exhaust gas flowing direction A. Each of the dielectrics 16 comprises an alumina (Al<sub>2</sub>O<sub>3</sub>) layer having a thickness of 0.5 mm, which is bonded to each of the second to sixth electrodes 14<sub>2</sub> to 14<sub>6</sub> by the mechanical pressing. The adhesion using an adhesive, the flame spraying or the like may be also utilized for such bonding. A gap  $g$  in each of the plasma spaces Pp, namely, a distance between each of the adjacent electrodes 14<sub>1</sub> to 14<sub>6</sub> and each of the dielectrics 16 is 0.5 mm.

A particulate material content reducing test was carried

out using the above-described equipment 1, according to the following procedure:

(1) The diesel generator 2 was operated, and an exhaust gas discharged from the diesel generator 2 was caused to flow through the exhaust gas 3 and the conduit 5.

(2) The flow rate of the testing exhaust gas flowing through the conduit 5, and hence through the plasma generating device PG, was regulated to 7.0 L/min by the first and second flow rate regulating valves 4<sub>1</sub> and 4<sub>2</sub>, while being measured by the flow-meter 8.

(3) The heater 6 was operated to heat the exhaust gas having a temperature of about 70°C up to 150°C, and the exhaust gas was allowed to flow through the first and third ports p<sub>1</sub> and p<sub>3</sub> of the three-way valve 10. Then, the testing exhaust gas flowing through the conduit 5 was diverted into the first branch pipe 9<sub>1</sub> by driving the suction pump 12 at a suction rate of 5.0 L/min, and caused to flow through the filter 13 over 10 minutes, whereby a particulate material in the testing exhaust gas was collected by the filter 13. A weight of the particulate material collected was determined from the weights of the filter before and after the collection of the particulate material, and defined as an amount of the particulate material before a plasma treatment.

(4) The plasma generating device PG was operated, and the second and third ports P2 and P3 were put into communication with each other by switching-over of the three-way valve 10. Then, the testing exhaust gas discharged from the plasma generating device PG and flowing through the conduit 5, was diverted into the second

branch pipe 9<sub>2</sub> by driving the suction pump 12 at a suction rate of 5.0 L/min, and caused to flow through a new filter 13 over 10 minutes, whereby the particulate material in the testing exhaust gas was collected by the new filter 13. A weight of the particulate material collected was determined from the weights of the filter before and after the collection of the particulate material, and defined as an amount of the particulate material after the plasma treatment.

In the test, an excitation spectrum of the exhaust gas was analyzed as a plasma excited state of the exhaust gas within the plasma generating device PG by an emission spectrometric photometer (IMUC-7000 made by Otsuka Electronics Co., Ltd.). As a result, the production of a plurality of excited-oxygen atoms by the generation of plasma was observed. Particularly, because an excitation spectrum of 6eV or more was present, the existence of O(<sup>1</sup>D) radicals was considered to be present. Thus, it can be said that a plurality of hydroxide excited species and per-hydroxide excited species HOO\* were produced.

Table 1 shows plasma generating conditions for Examples 1 to 3.

Table 1

θ <sub>max</sub>	Plasma space			Electricity input conditions			Intensity E of electric field (kV/mm)	Power density D <sub>w</sub> (W/cm <sup>3</sup> )
	Gap (mm)	Volume (cm <sup>3</sup> )	Alternate current	Frequency (Hz)	Voltage (kVp-p)	Power (W)		
1	0.5	2.5	Sine wave AC	300	3.0	2.7	3.0	1.1
2	1.5	7.5	Sine wave AC	700	8.2	9.1	2.7	1.2
3	0.5	2.5	Sine wave AC	100	3.0	2.1	3.0	0.8

In Table 1, the intensity E of electric field is a value provided by dividing a voltage  $[(kV_{p-p})/2]$  applied between the opposed electrodes such as between the first and second electrodes 14<sub>1</sub> and 14<sub>2</sub> by the gap  $g$  (mm), and the power density  $D_w$  is a value provided by dividing a power (0.54 W  $\times$  5 = 2.7 W) in the plasma space  $P_p$  defined by the opposed electrodes by a volume  $[(2 \text{ cm} \times 5 \text{ cm} \times 0.05 \text{ cm}) \times 5 = 2.5 \text{ cm}^3]$  of the plasma space  $P_p$ .

Table 2 shows amounts of the particulate material before and after the plasma treatment, and decrement amounts and decrement rates of the particulate material decreased by the plasma treatment for Examples 1 to 3.

Table 2

	Particulate material			
	Amount (mg) before plasma treatment	Amount (mg) after plasma treatment	Decrement amount (mg)	Decrement rate (%)
Example 1	1.3	0.2	1.1	84.6
Example 2	1.2	0.7	0.5	41.7
Example 3	1.4	0.8	0.6	42.9

Fig.3 is a graph based on Table 2 and showing amounts of the particulate material before and after the plasma treatment for Examples 1 to 3. As apparent from Tables 1 and 2 and Fig.3,

according to Example 1, per-hydroxide excited species  $\text{HO}^*$  can be efficiently produced by setting the intensity  $E$  of electric field at a value equal to or larger than 3.0 kV/mm and the power density  $D_w$  at a value equal to or larger than 1 W/cm<sup>3</sup>, whereby the decrement rate of the particulate material can be increased to about 84.6 %. The intensity  $E$  of electric field is preferably in a range of  $3 \text{ kV/mm} \leq E \leq 15 \text{ kV/mm}$ , and the power density  $D_w$  is preferably in a range of  $1 \text{ W/cm}^3 \leq D_w \leq 10 \text{ W/cm}^3$ .

In the cases of Examples 2 and 3, the decrement rate of the particulate material is lower and approximately half of that in Example 1, because one of the intensity  $E$  of electric field and the power density  $D_w$  dose not satisfies the above-described requirement.

Embodiment I is applicable for decreasing the content of a particulate material contained in an exhaust gas discharged from an automobile, a factory or the like.

According to the above-described process, it is possible to continuously convert the particulate material contained in the exhaust gas from the lean burn engine or the like by the oxidization, to thereby remarkably decrease the content thereof. In addition, this process cannot provide a disadvantage that the exhausting resistance is greatly increased to affect an engine output.

(Embodiment II)

In an equipment 1 for a particulate material content

decreasing test shown in Fig. 4, a commercially available diesel generator 2 is selected as a source of generating an exhaust gas containing a particulate material. A first flow rate regulating valve 4<sub>1</sub> is mounted in an exhaust pipe 3 of the diesel generator 2. In the exhaust pipe 3, a conduit 5 is connected at one end between the diesel generator 2 and the first flow rate regulating valve 4<sub>1</sub>, and a second flow rate regulating valve 4<sub>2</sub>, a heater 6, a catalytic reactor CR, a plasma generating device PG and a flow-meter 8 are mounted on the exhaust pipe 3 sequentially in the named order from the side of the exhaust pipe 3. The other end of the conduit 5 opens to the atmosphere.

In the conduit 5, a first branch pipe 9<sub>1</sub> is connected at one end between the heater 6 and the catalytic reactor CR, and at the other end to a suction port of a suction pump 12. A first on-off valve 20<sub>1</sub> and a filter 13 are mounted in the first branch pipe 9<sub>1</sub> sequentially from the side of the conduit 5. In the conduit 5, a second branch pipe 9<sub>2</sub> is also connected at one end between the catalytic reactor CR and the plasma generating device PG, and at the other end between the first on-off valve 20<sub>1</sub> and the filter 13 in the first branch pipe 9<sub>1</sub>. A second on-off valve 20<sub>2</sub> is mounted in the second branch pipe 9<sub>2</sub>. Further, in the conduit 5, a third branch pipe 9<sub>3</sub> is connected at one end between the plasma generating device PG and the flow-meter 8, and at the other end between a connection of the second branch pipe 9<sub>2</sub> and the filter 13 in the first branch pipe 9<sub>1</sub>. A third

on-off valve 20<sub>3</sub> is mounted in the third branch pipe 9<sub>3</sub>.

As in Embodiment I, the diesel generator 2 is EXT12D made by Honda Motor Co. Ltd., and the specification thereof is as follows: the type of an engine: a water-cooled 3-cylinder and 4-cycle diesel engine; a total displacement: 1061 cc; a fuel used: diesel light oil; a rated output: 12 kVA. The filter 13 is a Teflon-coated filter made by German Laboratory, and has a mesh size of 0.3  $\mu\text{m}$  mesh.

As shown in Fig.2, the plasma generating device PG has the same structure as that in Embodiment I and includes a plurality of first to sixth electrodes 14<sub>1</sub> to 14<sub>6</sub> made of a metal, each of which is formed into a plate shape in the illustrated embodiment. The first to sixth electrodes 14<sub>1</sub> to 14<sub>6</sub> are mounted in a housing 15 (see Fig.4) so that they are parallel to a direction A of flowing of an exhaust gas, and the adjacent electrodes 14<sub>1</sub> and 14<sub>2</sub>; 14<sub>2</sub> and 14<sub>3</sub>; 14<sub>3</sub> and 14<sub>4</sub>; 14<sub>4</sub> and 14<sub>5</sub>; 14<sub>5</sub> and 14<sub>6</sub> are opposed to each other. The surface of the first electrode 14<sub>1</sub> located at one end, which is opposed to the second electrode 14<sub>2</sub>, is a non-coated metal surface, but the surface of the second electrode 14<sub>2</sub>, which is opposed to the first electrode 14<sub>1</sub>, is coated entirely with a dielectric 16. The arrangement relationship between the opposed surfaces of the first and second electrodes 14<sub>1</sub> and 14<sub>2</sub> applies to those between the second and third electrodes 14<sub>2</sub> and 14<sub>3</sub>; between the third and fourth electrodes 14<sub>3</sub> and 14<sub>4</sub>; between the fourth and fifth

electrodes 14<sub>4</sub> and 14<sub>5</sub>; and between the fifth and sixth electrodes 14<sub>5</sub> and 14<sub>6</sub>. The first, third and fifth electrodes 14<sub>1</sub>, 14<sub>3</sub> and 14<sub>5</sub> are connected to a power source 18 through lead wires 17, while the second, fourth and sixth electrodes 14<sub>2</sub>, 14<sub>4</sub> and 14<sub>6</sub> are earthed through lead wires 19.

If the plasma generating device PG is constructed as described above, the entire surface of each of the dielectrics 16 is uniformly charged upon application of a voltage, and hence the entire space between each of the dielectrics 16 and each of the electrodes 14<sub>1</sub> to 14<sub>6</sub> which is opposed thereto constitutes a plasma space P<sub>p</sub>. Namely, the plasma spaces P<sub>p</sub> are formed between the first and second electrodes 14<sub>1</sub> and 14<sub>2</sub>; between the second and third electrodes 14<sub>2</sub> and 14<sub>3</sub>; between the third and fourth electrodes 14<sub>3</sub> and 14<sub>4</sub>; between the fourth and fifth electrodes 14<sub>4</sub> and 14<sub>5</sub>; and between the fifth and sixth electrodes 14<sub>5</sub> and 14<sub>6</sub>. That is, each of the plasma spaces P<sub>p</sub> is defined by the opposed electrodes.

Each of the first to sixth electrodes 14<sub>1</sub> to 14<sub>6</sub> is formed of a stainless steel (e.g., JIS SUS316), and has a size of 20 mm long, 50 mm wide and 1.0 mm thick, with a transverse side extending along the exhaust gas flowing direction A. Each of the dielectrics 16 comprises an alumina (Al<sub>2</sub>O<sub>3</sub>) layer having a thickness of 0.5 mm, which is bonded to each of the second to sixth electrodes 14<sub>2</sub> to 14<sub>6</sub> by the mechanical pressing. The adhesion using an adhesive, the flame spraying or the like may

be also utilized for such bonding. A gap  $g$  in each of the plasma spaces  $P_p$ , namely, a distance between each of the adjacent electrodes  $14_1$  to  $14_6$  and each of the dielectric  $16$  is 0.5 mm.

The catalytic reactor CR comprises a honeycomb carrier and platinum (Pt) carried as a catalyst on the honeycomb carrier, and is placed in a housing 21. To carry platinum on the honeycomb carrier, the following process was adopted: (i) 80 grams of a solution of platinum dinitro diamine nitric acid  $[Pt(NH_3)_2(NO_2)_2 \cdot HNO_3]$  containing 5 % by weight of platinum, 96 grams of commercially available  $\gamma$ - $Al_2O_3$ , and 1,000 grams of ion-exchange water were mixed together, and at the same time, an excessive amount of water was removed; (ii) a mixture was dried at 200°C for 2 hours; (iii) a powder of  $Al_2O_3$  carrying platinum was produced by calcining the mixture at 600°C for 2 hours; (iv) 90 grams of the powder of  $Al_2O_3$  carrying platinum, 50 grams of an  $SiO_2$  binder (containing 20 % by weight of  $SiO_2$ ) and 150 grams of ion-exchange water were mixed together; (v) a slurry was prepared by wet-pulverizing the mixture over 12 hours using alumina balls; (vi) a honeycomb carrier carrying a platinum catalyst in an amount of 4g/L at a wash coat amount of 100 g/L was prepared by repeating a process: immersing a honeycomb carrier made of cordierite and having a volume of about 30 L (a diameter of 2.54 and a length of 60 mm) with 400 cells/in<sup>2</sup> and 6 mils into the slurry; removing an excessive amount of the slurry from the carrier by jet water injection

after the removal of the carrier; and drying the carrier at 150°C for 1 hour; (vii) the honeycomb carrier was calcined at 500°C for 2 hours; (viii) the honeycomb carrier carrying the catalyst was placed into the housing 21.

This type of catalytic reactor CR has a structure substantially same as that of a catalytic converter, and hence the exhausting resistance cannot be greatly increased.

A particulate material content-decreasing test was carried out using the above-described equipment by the following procedure:

(1) The diesel generator 2 was operated, and an exhaust gas discharged from the diesel generator 2 was caused to flow through the exhaust gas 3 and the conduit 5.

(2) The flow rate of the testing exhaust gas flowing through the conduit 5, and hence through the catalytic reactor CR and the plasma generating device PG, was regulated to 7.0 L/min by the first and second flow rate regulating valves 4<sub>1</sub> and 4<sub>2</sub>, while being measured by the flow-meter 8.

(3) The heater 6 was operated to heat the exhaust gas having a temperature of about 70°C up to 150°C, and the first on-off valve 20<sub>1</sub> was opened, while the second and third on-off valves 20<sub>2</sub> and 20<sub>3</sub> were closed. Then, the testing exhaust gas flowing through the conduit 5 was diverted into the first branch pipe 9<sub>1</sub> by driving the suction pump 12 at a suction rate of 5.0 L/min, and caused to flow through the filter 13 over 20 minutes, whereby

a particulate material in the testing exhaust gas was collected by the filter 13. A weight of the particulate material collected was determined from the weights of the filter before and after the collection of the particulate material, and was defined as an amount of the particulate material before a catalytic reaction.

(4) The second on-off valve 20<sub>2</sub> was opened, while the first and third on-off valves 20<sub>1</sub> and 20<sub>3</sub> were closed. Then, the testing exhaust gas flowing through the conduit 5 via the catalytic reactor CR, was diverted into the second branch pipe 9<sub>2</sub> by driving the suction pump 12 at a suction rate of 5.0 L/min, and caused to flow through a new filter 13 over 20 minutes, whereby the particulate material in the testing exhaust gas was collected by the new filter 13. A weight of the particulate material collected was determined from the weights of the filter before and after the collection of the particulate material, and defined as an amount of the particulate material after the catalytic reaction.

(5) The plasma generating device PG was operated, and the third on-off valve 20<sub>3</sub> was opened, while the first and second on-off valves 20<sub>1</sub> and 20<sub>2</sub> were closed. Then, the testing exhaust gas flowing through the conduit 5 via the catalytic reactor CR and the plasma generating device PG, was diverted into the third branch pipe 93 by driving the suction pump 12 at a suction rate of 5.0 L/min, and caused to flow through a new filter 13 over

20 minutes, whereby the particulate material in the testing exhaust gas was collected by the new filter 13. A weight of the particulate material collected was determined from the weights of the filter before and after the collection of the particulate material, and defined as an amount of the particulate material after the plasma treatment.

The process using platinum as the catalyst as described above is defined as Example 4.

A particulate material content-decreasing test was carried out as Example 5 in a similar manner, except that a catalytic reactor CR having silver (Ag) as a catalyst was used, and the temperature of an exhaust gas flowing into the plasma generating device PG was set at 250°C. To carry silver (Ag) on  $\text{Al}_2\text{O}_3$  in a catalyst-carried honeycomb carrier, this process was the same as Example 3, except that a mixture comprising 4.72 grams of silver nitrate ( $\text{AgNO}_3$ ), 97 grams of commercially available  $\gamma\text{-Al}_2\text{O}_3$ , and 1,000 grams of ion-exchange water were used as a silver-containing solution, and the amount of silver carried on the honeycomb carrier was set at 3 g/L.

In each of Examples 4 and 5, an excitation spectrum of the exhaust gas was analyzed as a plasma excited state of the exhaust gas within the plasma generating device PG by an emission spectrometricphotometer (IMUC-7000 made by Otsuka Electronics Co., Ltd.). As a result, the production of a plurality of excited-oxygen atoms by the generation of plasma

was observed. Particularly, because an excitation spectrum of 6eV or more was present,  $O(^1D)$  radicals was considered to be present. Thus, it can be said that if the plasma is generated in the exhaust gas,  $O(^1D)$  radicals and ozone excited species are produced by the collision between electrons released from the electrodes and oxygen atoms  $\rightarrow$  hydroxide excited species  $OH^*$  are produced by the reaction of  $O(^1D)$  radicals and ozone excited species with water  $\rightarrow$  per-hydroxide excited species  $HOO^*$  having a strong oxidizing force are produced by the reaction of the hydroxide excited species  $OH^*$  with oxygen. Namely, the plasma excited species include  $O(^1D)$  radicals, hydroxide excited species  $OH^*$ , per-hydroxide excited species  $HOO^*$  and the like.

Table 3 shows plasma generating conditions for Examples 4 and 5.

Table 3

Example	Plasma space			Electricity input conditions			Intensity E of electric field (kV/mm)	Power density D <sub>w</sub> (W/cm <sup>3</sup> )
	Gap (mm)	Volume (cm <sup>3</sup> )	Alternate current	Frequency (Hz)	Voltage (kVp-p)	Power (W)		
4	0.5	2.5	Sine wave AC	2,500	7.6	7.4	7.6	3.0
5	0.5	2.5	Sine wave AC	1,600	7.2	9.5	7.2	3.8

In Table 3, the intensity  $E$  of electric field is a value provided by dividing a voltage  $[(kV_{p-p})/2]$  applied between the opposed electrodes such as between the first and second electrodes  $14_1$  and  $14_2$  by the gap  $g$  (mm), and the power density  $D_w$  is a value provided by dividing a power (Example 4:  $1.48 \text{ W} \times 5 = 7.4 \text{ W}$ ; and Example 5:  $1.9 \text{ W} \times 5 = 9.5 \text{ W}$ ) in the plasma space  $P_p$  defined by the opposed electrodes by a volume  $[(2 \text{ cm} \times 5 \text{ cm} \times 0.05 \text{ cm}) \times 5 = 2.5 \text{ cm}^3]$  of the plasma space  $P_p$ .

Table 4 shows amounts of the particulate material before and after the catalytic reaction and after the plasma treatment, and decrement amounts and decrement rates of the particulate material decreased by the catalytic reaction and the plasma treatment for Examples 4 and 5.

Table 4

Amount before catalytic reaction (mg)	After catalytic reaction			After plasma treatment		
	Decrement amount (mg)	Decrement rate (%)	Amount (mg)	Decrement amount (mg)	Decrement rate (%)	
4	2.6	1.9	0.7	26.9	0.2	2.4
5	2.4	1.7	0.7	29.2	0.2	2.2

Figs.5 and 6 are graphs based on Table 4 and showing amounts of the particulate material before and after the catalytic reaction and after the plasma treatment for Examples 4 and 5. As apparent from Tables 3 and 4 and Figs.5 and 6, according to Examples 4 and 5, plasma excited species can be efficiently produced by setting the intensity  $E$  of electric field at a value equal to or larger than 3.0 kV/mm and setting the power density  $D_w$  at a value equal to or larger than 1 W/cm<sup>3</sup>, whereby the decrement rate of the particulate material can be increased up to about 92 % or more. The intensity  $E$  of electric field is preferably in a range of  $3 \text{ kV/mm} \leq E \leq 15 \text{ kV/mm}$ , and the power density  $D_w$  is preferably in a range of  $1 \text{ W/cm}^3 \leq D_w \leq 10 \text{ W/cm}^3$ .

The particulate material before the catalytic reaction was subjected to an extraction using a Soxhlet extractor. As a result, an amount of a fraction soluble in an organic solvent was about 30 % by weight, and an amount of carbon solid fraction was about 70 % by weight. If Table 4 is reviewed with this fact in mind, it can be seen that substantially all the fraction soluble in the organic solvent was converted for purification of the exhaust gas by the catalytic reaction, and the conversion of the carbon solid fraction for purification of the exhaust gas was efficiently carried out by the subsequent plasma treatment.

Embodiment II is applicable for decreasing the content of a particulate material contained in an exhaust gas discharged from an automobile, a factory or the like.

According to the above-described process, it is possible to continuously convert a particulate material contained in an exhaust gas from a lean burn engine or the like by the oxidization, to thereby remarkably decrease the content of the particulate material. In addition, this process cannot provide a disadvantage that the exhausting resistance is greatly increased to affect an engine output.

(Embodiment III)

In an equipment 1 for a particulate material content-decreasing test shown in Fig. 7, a commercially available diesel generator 2 is selected as a source of generating an exhaust gas containing a particulate material. A first flow rate regulating valve 4<sub>1</sub> is mounted in an exhaust pipe 3 of the diesel generator 2. In the exhaust pipe 3, a conduit 5 is connected at one end between the diesel generator 2 and the first flow rate regulating valve 4<sub>1</sub>, and a second flow rate regulating valve 4<sub>2</sub>, a heater 6, a plasma generating device PG, a catalytic reactor CR and a flow-meter 8 are mounted on the exhaust pipe 3 sequentially in the named order from the side of the exhaust pipe 3. The other end of the conduit 5 opens to the atmosphere.

In the conduit 5, a first branch pipe 9<sub>1</sub> is connected at one end between the heater 6 and the plasma generating device

PG, and at the other end to a suction port of a suction pump 12. A first on-off valve 20<sub>1</sub> and a filter 13 are mounted in the first branch pipe 9<sub>1</sub> sequentially from the side of the conduit 5. In the conduit 5, a second branch pipe 9<sub>2</sub> is also connected at one end between the plasma generating device PG and the catalytic reactor CR, and at the other end between the first on-off valve 20<sub>1</sub> and the filter 13 in the first branch pipe 9<sub>1</sub>. A second on-off valve 20<sub>2</sub> is mounted in the second branch pipe 9<sub>2</sub>. Further, in the conduit 5, a third branch pipe 9<sub>3</sub> is connected at one end between the catalytic reactor CR and the flow-meter 8, and at the other end between a connection of the second branch pipe 9<sub>2</sub> and the filter 13 in the first branch pipe 9<sub>1</sub>. A third on-off valve 20<sub>3</sub> is mounted in the third branch pipe 9<sub>3</sub>.

As in Embodiments I and II, the diesel generator 2 is EXT12D made by Honda Motor Co. Ltd., and the specification thereof is as follows: the type of an engine: a water-cooled 3-cylinder and 4-cycle diesel engine; a total displacement: 1061 cc; a fuel used: diesel light oil; a rated output: 12 kVA. The filter 13 is a Teflon-coated filter made by German Laboratory, and has a mesh size of 0.3  $\mu\text{m}$  mesh.

As shown in Fig.2, the plasma generating device PG has the same structure as that in Embodiments I and II, and includes a plurality of first to sixth electrodes 14<sub>1</sub> to 14<sub>6</sub> made of a metal, each of which is formed into a plate shape in the

illustrated embodiment. The first to sixth electrodes 14<sub>1</sub> to 14<sub>6</sub> are mounted in a housing 15 (see Fig. 7) so that they are parallel to a direction A of flowing of an exhaust gas, and the adjacent electrodes 14<sub>1</sub> and 14<sub>2</sub>; 14<sub>2</sub> and 14<sub>3</sub>; 14<sub>3</sub> and 14<sub>4</sub>; 14<sub>4</sub> and 14<sub>5</sub>; 14<sub>5</sub> and 14<sub>6</sub> are opposed to each other. The surface of the first electrode 14<sub>1</sub> located at one end, which is opposed to the second electrode 14<sub>2</sub>, is a non-coated metal surface, but the surface of the second electrode 14<sub>2</sub>, which is opposed to the first electrode 14<sub>1</sub>, is coated entirely with a dielectric 16. The arrangement relationship between the opposed surfaces of the first and second electrodes 14<sub>1</sub> and 14<sub>2</sub> applies to those between the second and third electrodes 14<sub>2</sub> and 14<sub>3</sub>; between the third and fourth electrodes 14<sub>3</sub> and 14<sub>4</sub>; between the fourth and fifth electrodes 14<sub>4</sub> and 14<sub>5</sub>; and between the fifth and sixth electrodes 14<sub>5</sub> and 14<sub>6</sub>. The first, third and fifth electrodes 14<sub>1</sub>, 14<sub>3</sub> and 14<sub>5</sub> are connected to a power source 18 through lead wires 17, while the second, fourth and sixth electrodes 14<sub>2</sub>, 14<sub>4</sub> and 14<sub>6</sub> are earthed through lead wires 19.

If the plasma generating device PG is constructed as described above, the entire surface of each of the dielectrics 16 is uniformly charged upon application of a voltage, and hence the entire space between each of the dielectrics 16 and each of the electrodes 14<sub>1</sub> to 14<sub>6</sub> which is opposed thereto constitutes a plasma space Pp. Namely, the plasma spaces Pp are formed between the first and second electrodes 14<sub>1</sub> and 14<sub>2</sub>; between

the second and third electrodes 14<sub>2</sub> and 14<sub>3</sub>; between the third and fourth electrodes 14<sub>3</sub> and 14<sub>4</sub>; between the fourth and fifth electrodes 14<sub>4</sub> and 14<sub>5</sub>; and between the fifth and sixth electrodes 14<sub>5</sub> and 14<sub>6</sub>. That is, each of the plasma spaces P<sub>p</sub> is defined by the opposed electrodes.

Each of the first to sixth electrodes 14<sub>1</sub> to 14<sub>6</sub> is formed of a stainless steel (e.g., JIS SUS316), and has a size of 20 mm long, 50 mm wide and 1.0 mm thick, with a transverse side extending along the exhaust gas flowing direction A. Each of the dielectrics 16 comprises an alumina (Al<sub>2</sub>O<sub>3</sub>) layer having a thickness of 0.5 mm, which is bonded to each of the second to sixth electrodes 14<sub>2</sub> to 14<sub>6</sub> by the mechanical pressing. The adhesion using an adhesive, the flame spraying or the like may be also utilized for such bonding. A gap g in each of the plasma spaces P<sub>p</sub>, namely, a distance between each of the adjacent electrodes 14<sub>1</sub> to 14<sub>6</sub> and each of the dielectric 16 is 0.5 mm.

The catalytic reactor CR comprises a honeycomb carrier and platinum (Pt) carried as a catalyst on the honeycomb carrier, and is placed in a housing 21. To carry platinum on the honeycomb carrier, the following process was adopted: (i) 80 grams of a solution of platinum dinitrodiamine nitric acid [Pt(NH<sub>3</sub>)<sub>2</sub>(NO<sub>2</sub>)<sub>2</sub>•HNO<sub>3</sub>] containing 5 % by weight of platinum, 96 grams of commercially available  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, and 1,000 grams of ion-exchange water were mixed together, and at the same time, an excessive amount of water was removed; (ii) a mixture was

dried at 200°C for 2 hours; (iii) a powder of Al<sub>2</sub>O<sub>3</sub> carrying platinum was produced by calcining the mixture at 600°C for 2 hours; (iv) 90 grams of the powder of Al<sub>2</sub>O<sub>3</sub> having platinum carried thereon, 50 grams of an SiO<sub>2</sub> binder (containing 20 % by weight of SiO<sub>2</sub>) and 150 grams of ion-exchange water were mixed together; (v) a slurry was prepared by wet-pulverizing the mixture over 12 hours using alumina balls; (vi) a honeycomb carrier carrying a platinum catalyst in an amount of 4g/L at a wash coat amount of 100 g/L was prepared by repeating a process: immersing a honeycomb carrier made of cordierite and having a volume of about 30 L (a diameter of 2.54 and a length of 60 mm) with 400 cells/in<sup>2</sup> and 6 mils into the slurry; removing an excessive amount of the slurry from the carrier by jet water injection after the removal of the carrier; and drying the carrier at 150°C for 1 hour; (vii) the honeycomb carrier was calcined at 500°C for 2 hours; (viii) the honeycomb carrier carrying the catalyst was placed into the housing 21.

This type of catalytic reactor CR has a structure substantially same as that of a catalytic converter, and hence the exhausting resistance cannot be greatly increased.

A particulate material content-decreasing test was carried out using the above-described equipment by the following procedure:

(1) The diesel generator 2 was operated, and an exhaust gas discharged from the diesel generator 2 was caused to flow

through the exhaust gas 3 and the conduit 5.

(2) The flow rate of the testing exhaust gas flowing through the conduit 5, and hence through the plasma generating device PG and the catalytic reactor CR, was regulated to 7.0 L/min by the first and second flow rate regulating valves 4<sub>1</sub> and 4<sub>2</sub>, while being measured by the flow-meter 8.

(3) The heater 6 was operated to heat the exhaust gas having a temperature of about 70°C up to 150°C, and the first on-off valve 20<sub>1</sub> was opened, while the second and third on-off valves 20<sub>2</sub> and 20<sub>3</sub> were closed. Then, the testing exhaust gas flowing through the conduit 5 was diverted into the first branch pipe 9<sub>1</sub> by driving the suction pump 12 at a suction rate of 5.0 L/min, and caused to flow through the filter 13 over 20 minutes, whereby a particulate material in the testing exhaust gas was collected by the filter 13. A weight of the particulate material collected was determined from the weights of the filter before and after the collection of the particulate material, and was defined as an amount of the particulate material before a catalytic reaction.

(4) The plasma generating device PG was operated, and the second on-off valve 20<sub>2</sub> was opened, while the first and third on-off valves 20<sub>1</sub> and 20<sub>3</sub> were closed. Then, the testing exhaust gas discharged from the plasma generating device PG and flowing through the conduit 5 via the catalytic reactor CR, was diverted into the second branch pipe 9<sub>2</sub> by driving the suction pump 12

at a suction rate of 5.0 L/min, and caused to flow through a new filter 13 over 20 minutes, whereby the particulate material in the testing exhaust gas was collected by the new filter 13. A weight of the particulate material collected was determined from the weights of the filter before and after the collection of the particulate material, and defined as an amount of the particulate material after the plasma treatment.

(5) In a state in which the plasma generating device PG was operated, the third on-off valve 20<sub>3</sub> was opened, while the first and second on-off valves 20<sub>1</sub> and 20<sub>2</sub> were closed. Then, the testing exhaust gas flowing through the conduit 5 via the plasma generating device PG and the catalytic reactor CR, was diverted into the third branch pipe 93 by driving the suction pump 12 at a suction rate of 5.0 L/min, and caused to flow through a new filter 13 over 20 minutes, whereby the particulate material in the testing exhaust gas was collected by the new filter 13. A weight of the particulate material collected was determined from the weights of the filter before and after the collection of the particulate material, and defined as an amount of the particulate material after the catalytic reaction.

The process using platinum as the catalyst as described above is defined as Example 6.

A particulate material content-decreasing test was carried out as Example 7 in a similar manner, except that a catalytic reactor CR having silver (Ag) as a catalyst was used,

and the temperature of an exhaust gas flowing into the plasma generating device PG was set at 250°C. To carry silver (Ag) on Al<sub>2</sub>O<sub>3</sub> in a catalyst-carried honeycomb carrier, this process was the same as Example 6, except that a mixture comprising 4.72 grams of silver nitrate (AgNO<sub>3</sub>), 97 grams of commercially available  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, and 1,000 grams of ion-exchange water were used as a silver-containing solution, and the amount of silver carried on the honeycomb carrier was set at 3 g/L.

In each of Examples 6 and 7, the exhaust gas upstream and the exhaust gas downstream of the plasma generating device PG was sampled, and the type of the gas was analyzed by FT-IR (a Fourier transform infrared spectroscopic device). As a result, the production of a plurality of nitrogen dioxide molecules and a plurality of ozone molecules by the generation of plasma was observed.

Table 5 shows plasma generating conditions for Examples 6 and 7.

Table 5

Examp E <sub>1</sub>	Plasma space			Electricity input conditions			Intensity E of electric field (kV/mm)	Power density D <sub>w</sub> (W/cm <sup>3</sup> )
	Gap (mm)	Volume (cm <sup>3</sup> )	Alternate current	Frequency (Hz)	Voltage (kVp-p)	Power (W)		
6	0.5	2.5	Sine wave AC	200	7.6	3.1	7.6	1.2
7	0.5	2.5	Sine wave AC	500	6.6	8.5	6.6	3.4

In Table 5, the intensity E of electric field is a value provided by dividing a voltage  $[(kV_{p-p})/2]$  applied between the opposed electrodes such as between the first and second electrodes 14<sub>1</sub> and 14<sub>2</sub> by the gap  $g$  (mm), and the power density  $D_w$  is a value provided by dividing a power (Example 6: 0.62 W  $\times$  5 = 3.1 W; and Example 7: 1.7 W  $\times$  5 = 8.5 W) in the plasma space  $P_p$  defined by the opposed electrodes by a volume [(2 cm  $\times$  5 cm  $\times$  0.05 cm)  $\times$  5 = 2.5 cm<sup>3</sup>] of the plasma space  $P_p$ .

Table 6 shows amounts of the particulate material before and after the plasma treatment and after the catalytic reaction, and decrement amounts and decrement rates of the particulate material decreased by the plasma treatment and the catalytic reaction for Examples 6 and 7.

Table 6

Example	Particulate material				
	After plasma treatment	Decrement amount (mg)	Decrement rate (%)	Amount (mg)	Decrement amount (mg)
6	2.5	0.4	2.1	84.0	0.1
7	2.3	0.3	2.0	87.0	0.1

Figs.8 and 9 are graphs based on Table 6 and showing amounts of the particulate material before and after the plasma treatment and after the catalytic reaction for Examples 6 and 7. As apparent from Tables 5 and 6 and Figs.8 and 9, according to Examples 6 and 7, nitrogen dioxide and ozone can be efficiently produced by setting the intensity E of electric field at a value equal to or larger than 3.0 kV/mm and setting the power density Dw at a value equal to or larger than 1 W/cm<sup>3</sup>, whereby the decrement rate of the particulate material can be increased up to about 84 % or more. The intensity E of electric field is preferably in a range of 3 kV/mm  $\leq$  E  $\leq$  15 kV/mm, and the power density Dw is preferably in a range of 1 W/cm<sup>3</sup>  $\leq$  Dw  $\leq$  10 W/cm<sup>3</sup>. Further, the decrement rate of the particulate material can be further increased up to approximately 96 % by carrying out the catalytic reaction.

Embodiment III is applicable for decreasing the content of a particulate material contained in an exhaust gas discharged from an automobile, a factory or the like.

According to the above-described process, it is possible to continuously convert a particulate material contained in an exhaust gas from a lean burn engine or the like by the oxidization, to thereby remarkably decrease the content of the particulate material. In addition, this process cannot provide a disadvantage that the exhausting resistance is greatly increased to affect an engine output.